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Enhancing the mechanical properties of natural jute yarn suitable for structural applications

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Abstract

Manufacturing natural-based high-performance composites are becoming of greater interest to the composite manufacturers and to their end-users due to their bio-degradability, low cost and availability. Yarn based textile architecture is commonly used in manufacturing these composites due to their excellent formability. However, for using natural based yarn as a reinforcing architecture in high load-bearing structural composite applications, a significant improvement in mechanical performance is required. Particularly, jute fibre yarn suffers from poor mechanical properties due to the presence of a fibrillar network, polysaccharides and other impurities in the fibre. For achieving this, we use aqueous glycine treatment (10%, W/V) on alkali(0.5 %, W/V) and untreated jute yarns for the first time. The glycine treatment on alkali-treated jute yarns (ATG) shows a huge improvement in tensile strength and strain values by almost ~105% and ~50 % respectively compared to untreated jute yarns (UT) because of the strong interactions and bonds developed between glycine, alkali and jute yarns. It is believed that the newly developed glycine treated jute yarns will be helpful to promote jute yarns in composite industries where load-bearing is the primary requirement and replace their synthetic counterparts.

1. Introduction

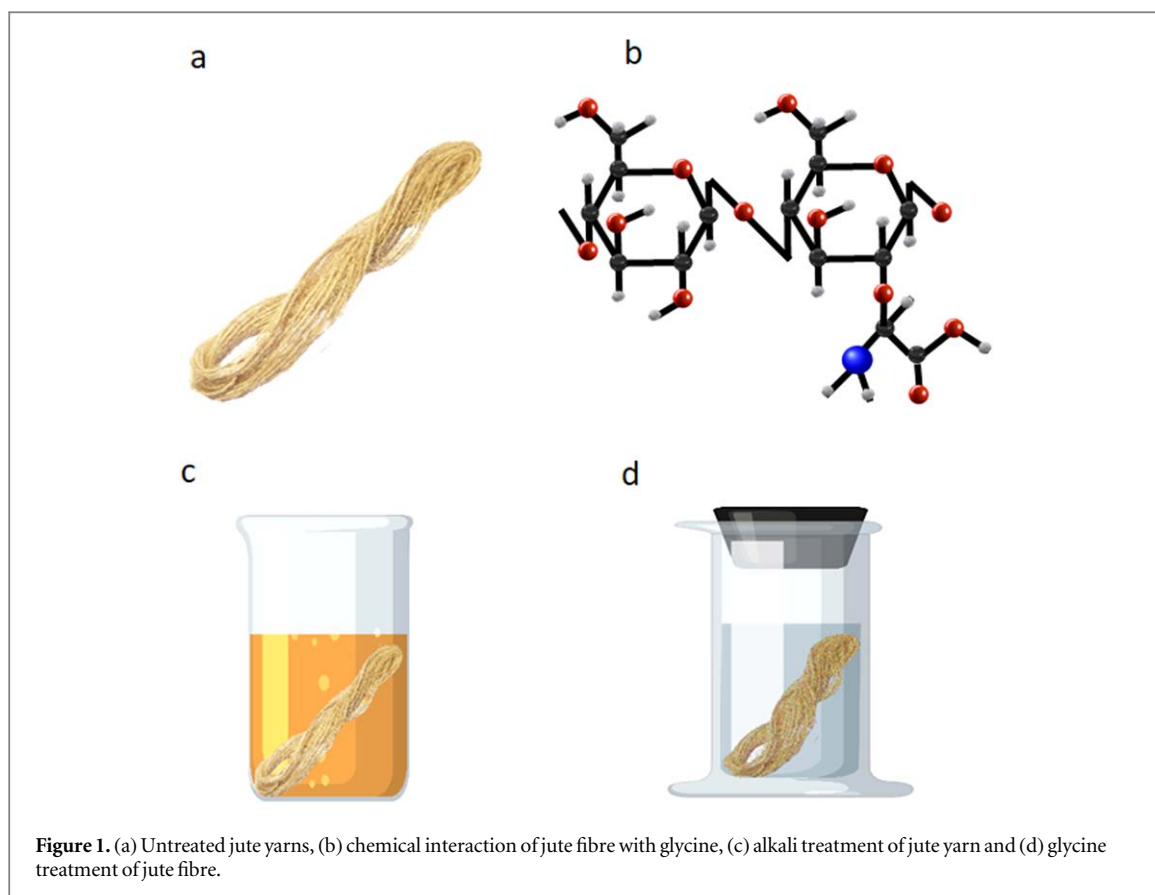
Fibre-reinforced composites have gained significant interests in recent years, due to their design flexibility, durability, chemical resistance, and relatively higher strength and stiffness at a low weight ratio. Traditionally, fibre reinforced composites are composed of synthetic fibres including glass, carbon and aramid as reinforcements in a polymer matrix [1]. However, synthetic fibres are not environmentally friendly as they are manufactured from fossil fuels, associated with relatively higher energy consumption and carbon emission. Natural fibres reinforced composites can be an environmentally sustainable alternative to their synthetic counterpart, due to lower environmental impacts including less carbon emission, less energy consumption and biodegradability [2–5]. The most promising natural plant fibres are jute, hemp, ramie, sisal, flax, and bamboo which could potentially replace synthetic fibres for various applications. Arguably, jute is the most attractive alternative amongst other natural plant fibres, due to its abundance, low production cost, lower density and high individual fibre length, as well as reasonable mechanical properties [5, 6]. In addition, Jute is the second most-produced natural fibre in the world after cotton (~3.63 million tons), and at least ~50% cheaper than flax and

other similar natural fibres. The use of jute for various applications could boost the farming economies of developing countries such as Bangladesh and India, where it is mostly produced.

The mechanical properties of jute are related to its relatively higher degree of crystallinity (~58%), and higher cellulose content (~70%) (table S1, Supporting Information (available online at stacks.iop.org/MRX/8/055503/mmedia)) [7]. Jute fibres have a 'fringed fibril model', where the inter-fibrillar matrix contains hemicellulose within ultimate cells and the middle lamella contains lignin between ultimate cells [8]. The viscoelastic and tensile behaviour of jute fibre depends on such polysaccharides (e.g. hemicellulose, lignin and pectin) and their relative proportion because they create links between the cellulosic microfibrils and are responsible for stress transfer among them [9]. It has been reported in the previous study that the removal of polysaccharides eliminates the microvoids present between the ultimate cell and middle lamella of the fibre. As a result, microfibrils present in the fibre become more parallel and homogeneity of the fibre improves which results in improvement of the failure stress, failure strain and stiffness of jute fibres [9]. The alkali treatment is the most commonly used method to remove polysaccharides i.e. hemicellulose, lignin, pectin, which improves the load-bearing capacity of jute fibre as a reinforcing material for fibre reinforced composites [6, 9]. However, jute fibre still contains micro-voids among the fibrils which limit their load-bearing capacity and create a weak fibre/matrix interface [6]. Previous studies have reported the removal of such defects *via* chemical [10–12], physical [13] and nanomaterials modifications [5, 13, 14] of jute fibres, which are time-consuming and expensive. Additionally, there are concerns with nanomaterials safety and their potential carcinogenic nature to health. The alkali treatment is the commonly used chemical medication for natural fibres as it removes surface waxes and affects hemicelluloses. The alkali treatment (0.5 wt.-%) of jute fibres with the prolonged exposure into the alkali solution is considered to be the most effective way of removing hemicelluloses without affecting lignins significantly [5, 15]. Further to alkali treatment, it is necessary to modify jute fibre to remove the flaws (microvoids) due to removal of lignins from the skin of fibre may also generate stress concentration. As a result, further chemical modification is necessary to solve these issues. However, the treatment should be cost-effective, environmentally friendly and easy to scale up. At present nano surface modification is becoming popular in modifying natural fibres in composites applications. However, dealing with these materials are health hazardous and costly.

For fibre-reinforced composites (FRC), yarn-based multi-axial textile architectures offer better mechanical properties including impact, compression after impact damage and interfacial strength, than the most popular unidirectional yarn architectures [16–19]. Textile architectures are mainly induced with plain, twill, sateen, and knitted derivatives which are mainly manufactured from yarns. Therefore, there exists a growing demand for FRC composite preforms comprised of woven fabric with multi-axial yarn architectures. However, the use of the jute yarn for structural FRC applications is limited due to its poor performance properties. The tensile strength for jute yarn was reported ~42–45 MPa only. Such a lower tensile strength is mainly due to the fibre impurities and the twist imparted to the fibre during spinning into yarn [20, 21]. In addition, the strain to failure of jute yarn was found to be limited ~6.0%–7.5% with a large scattering in the value in those studies [19, 20]. Recently, few studies have been carried out on the nano-modification of natural jute yarns to increase the strength and interfacial performance of the composites [21, 22]. It was found that the lower value of strain % is responsible for the amorphous phase's viscoelastic shear deformation present in the cellulose of the fibre [23]. Such viscoelastic shear deformation can be avoided by improving the crystallinity of fibres. In addition, the tensile strength can be improved by aligning the microfibrils in parallel directions with the loading axis [10, 24]. However, the improvement of mechanical properties of natural jute yarns are still limited, and not yet well understood. Nevertheless, it is well established that the deformation of natural fibres largely depends on the interphase of elementary fibres in the yarn. Recently, glycine-based protein materials have been used in cellulosic materials to improve strain to failure and wettability of cotton fibres. Glycine was first applied on cotton fibres to improve tensile properties. It was found that the glycine treatment improved the strain% of fibres by 36% [25]. This is mainly due to the interactions between the amino functional group of glycine and carboxylic groups present in the amorphous region of the cellulosic fibres that creates a strong chemical bond; thus, improves the mechanical properties of cotton fibres. As jute has similar cellulosic structures to cotton, glycine is believed to have similar effects to improve the mechanical properties of Jute fibres, which has not been reported yet to the best of our knowledge. In addition, glycine is an environmentally friendly simplest form of protein-based amino acid that has been extensively used in the drug industry. The low price and nonhazardous feature of glycine and the presence of amino-functional group have widened the scope of using this material in many applications [26].

Here, we report for the first time the improvement of tensile properties of jute yarns including the tensile strength and strain via a combination of alkali and aqueous glycine treatment. We optimize the processing conditions and parameters for such treatment to achieve the best mechanical properties of jute yarns. The untreated jute fibres were treated with a lower concentration (0.5 wt.-%) of alkali treatment, followed by a subsequent glycine treatment on the alkali-treated jute yarns. Surface topography of the treated and untreated jute yarns was examined using optical and scanning electron microscopes (SEM). In addition, changes in the



diameter of the yarns were recorded using an optical microscope, tensile properties were assessed *via* single yarn testing for 50 samples. We used Weibull statistics to analyse fibre failures after tensile tests. Chemical and thermal analysis were performed using Fourier transform infrared spectroscopy (FTIR), X-ray photoelectron spectroscopy (XPS) and Thermogravimetric analysis (TGA), respectively.

2. Materials and methods

2.1. Materials

The jute yarns were kindly donated by UMC Jute Mills Ltd, Narsinghdi, Bangladesh. Jute (*Corchorus Olitorius*) plants were harvested in Bangladesh *via* traditional methods and then processed in jute mills. The fibres used in the manufacturing of jute yarn were pretreated with vegetable oil to fibrillate the technical jute fibres to improve the spinnability of the fibre (figures 1(a) and S1(a), Supporting Information). As supplied jute yarns have a linear density of ~ 9.5 lbs/spindle and a TPI of ~ 4.09 (Table S2, Supporting Information). Sodium hydroxide (EMSURE[®] ISO, $>99\%$) pellets were supplied by Merck (Germany). Glycine (Amino Acetic Acid, $\text{NH}_2\text{CH}_2\text{COOH}$ 99.88% a white crystalline powder, Laboratory Reagent LR grade) used in this study was supplied by Fine Chemical Industries, India.

2.2. Alkali treatment

The untreated jute yarn (UT) were treated with 0.25%, 0.5%, 1% NaOH at room temperature for 24 h, and 2% NaOH at room temperature for 2 h at material to liquor ratio (M: L) of 1:30 (figures 1(c) and S1(b), Supporting Information). The optimized alkali concentration was selected based on the results obtained from the tensile test of alkali-treated fibres, which was then used for glycine treatment. After alkali treatment, all samples were thoroughly washed with distilled water until all the alkali traces were removed from treated yarns. Then all the samples were air-dried overnight at 50°C in an oven. The optimized alkali-treated fibres were labelled as AT.

2.3. Glycine treatment

Alkali treated (AT) and untreated jute yarns were treated with aqueous glycine at different concentrations (5, 10, 15 and 20 g l^{-1}) at 100°C and pH 7 for 1.5 h with an M:L ratio of 1:20 in an infrared lab dyeing machine (figures 1(d) and S2(a)–(b), Supporting Information). In addition, four pH levels (3, 5, 7 and 11) were also selected for 10 g l^{-1} aqueous solutions of glycine to see their effect on alkali-treated jute yarns. All of the treated

samples were thoroughly washed with distilled water and dried at 50 °C for at least 5 h in the oven. Here, untreated jute yarn with glycine treatment is labelled as UTG, while alkali-treated jute yarn with glycine treatment was identified as ATG.

2.4. Optical and scanning electron microscope (SEM)

An optical microscope (Jiusion Original 40-1000X, China) was used to observe the fibre surface at different magnifications, and measurement of the yarn diameter (figure S3, Supporting Information). A Scanning Electron Microscope (SEM, Model-SU 1510, Brand- Hitachi, Japan) was used to characterize the fibrillar packing of jute yarn. The samples were observed under SEM without any further coating on their surface using a low accelerating voltage (~2 kV).

2.5. Chemical and thermal characterization

The surface chemical composition of untreated and treated jute yarns were analyzed using a Kratos axis X-ray photoelectron spectroscopy (XPS) and a Fourier transform infrared spectroscopy (FTIR). The thermal decomposition of untreated and glycine treated jute yarn was analyzed using a TA instrument (TGA Q50, UK) from room temp to 600 °C in a nitrogen atmosphere at a 10 °C/min heating rate.

2.6. Tensile test

For the tensile testing, yarn samples were taken randomly from the spool of alkali (UT, AT) and glycine-treated (UTG, ATG) jute yarns, and conditioned in a standard laboratory atmosphere (65% relative humidity and 20 ± 2 °C) for 24 h before the final testing. A Universal Strength Tester (Testometric Model-M250-3CT, UK), was used with a load cell capacity of 25 KG to measure the breaking force and elongation at break of jute yarn according to the ASTM D2256-01 standard method. Single yarns were tested using a 50 mm gauge length at a cross-head speed of 2 mm/min as reported in the previous work [27]. The yarn was set on the machine using a special pneumatic yarn gripper supplied by Testometric, which ensures no slippage during testing (figures S4(a)–(b), Supporting Information). The tensile modulus of the yarns was calculated from the slope of 0.1%–0.3% strain of the yarn.

2.7. Weibull statistical analysis

The tensile properties of natural fibres (strength, modulus and strain %) are often described by the weakest link theory which is based on the statement that the materials are made of small elements and the elements are linked together. A material is considered to have failed if any of these small elements have failed [28]. The cumulative probability of failure for tensile and interface properties are given by the following formula:

$$P = 1 - \exp\left(-\left(\frac{\sigma}{\sigma_o}\right)^m\right) \quad (1)$$

$$P = 1 - \exp\left(-\left(\frac{E}{E_o}\right)^m\right) \quad (2)$$

$$P = 1 - \exp\left(-\left(\frac{\varepsilon_s}{\varepsilon_o}\right)^m\right) \quad (3)$$

where, σ is the tensile strength; σ_o is the Weibull scale parameter and m is the Weibull shape parameter. Similarly, E is the tensile modulus of the fibre; E_o is the Weibull scale parameter and m is the Weibull shape modulus, where, ε_s is the tensile strain, ε_o is the Weibull scale parameter and m is the Weibull shape parameter. Based on equations (1)–(3) a double natural logarithm is taken on both sides, which is shown in equations (4)–(6):

$$\ln(-\ln(1 - P)) = m \ln(\sigma - m \ln \sigma_o) \quad (4)$$

$$\ln(-\ln(1 - P)) = m \ln(E - m \ln E_o) \quad (5)$$

$$\ln(-\ln(1 - P)) = m \ln(\varepsilon - m \ln \varepsilon_o) \quad (6)$$

Where the cumulative probability of failure is related to the tensile strength (σ), tensile modulus (E), tensile strain (ε), the Weibull modulus (m) and σ_o , E_o and ε_o are the scale parameters of strength, the tensile modulus of the fibre and tensile strain of the yarn.

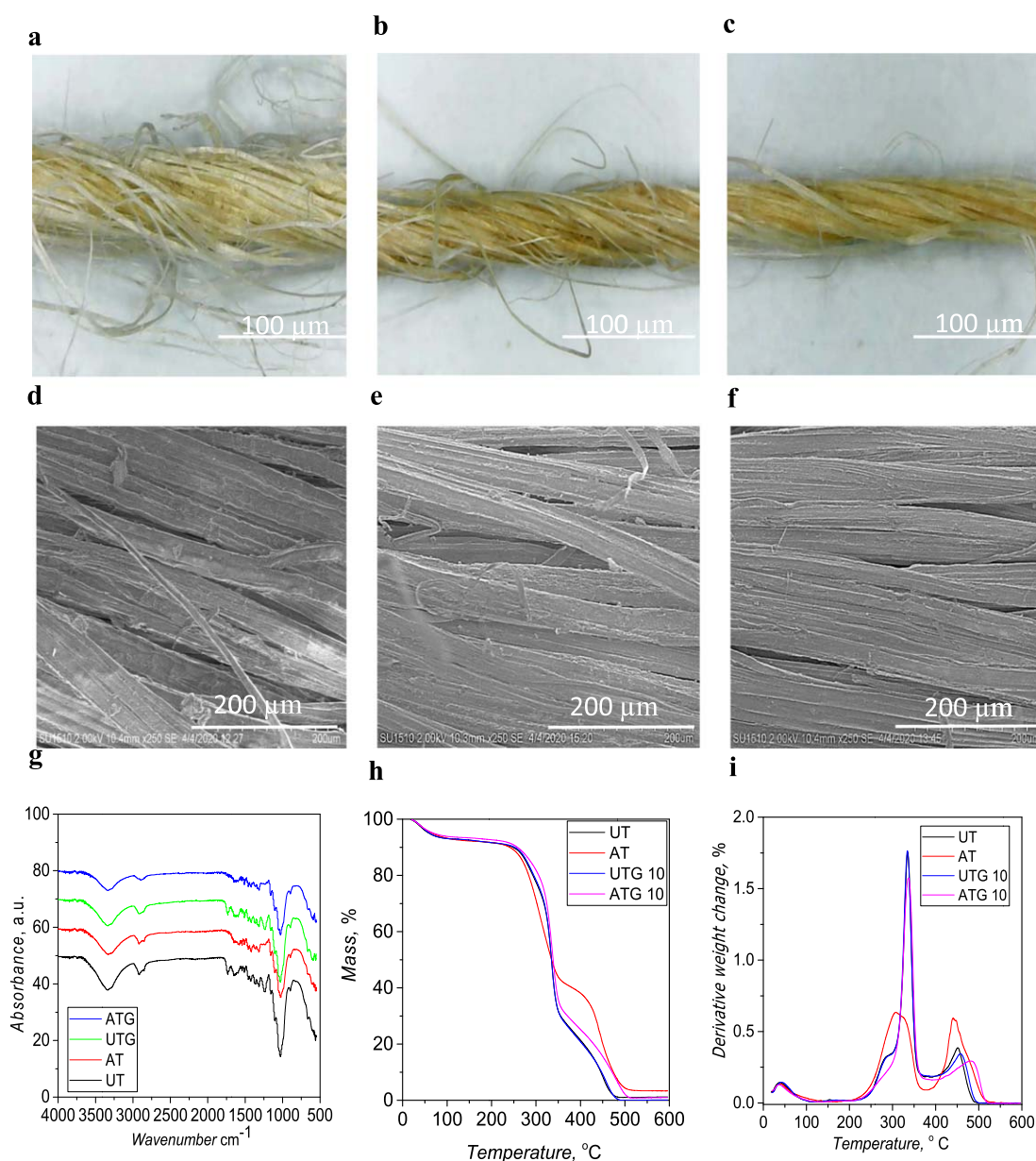


Figure 2. (a) Optical micrograph of UT jute yarn (X20), (b) Optical micrograph of AT jute yarn (X20), (c) Optical micrograph of ATG jute yarn (X20), (d) SEM micrograph of UT jute yarn (X250), (e) SEM micrograph of AT jute yarn (X250), (f) SEM micrograph of ATG jute yarn (X250), (g) FTIR spectra of untreated and treated jute yarns, (h) TGA curves of untreated and treated jute yarns and (i) Derivation weight % Vs temperature profile curves of untreated and treated yarn.

3. Results and discussion

3.1. Surface morphology

Figures 2(a)–(c) show optical images of untreated and treated jute yarns (figure S5a, Supporting Information). UT jute yarns show a loose appearance of technical fibres in the yarn, figure 2(a). The presence of impurities such as hemicelluloses and lignins in the interfibrillar network of fibres increase fibre irregularity may cause the loose appearance of fibres in the yarn.

However, after the alkali treatment, a noticeable change in the fibre packing of AT yarn (figure 2(b)) was observed which could be related to the removal of hemicelluloses after alkali treatment. As a result, yarn diameters were slightly reduced from ~ 0.92 mm to ~ 0.71 mm (Table S4, Supporting Information), which is in agreement with the results obtained in previous studies [6, 28, 29]. Glycine treatment on untreated jute fibre (UTG) slightly improved the packing of technical fibre in jute yarn while no change in the diameter was observed for UTG jute yarn (figure S5(a) and Table S4, Supporting Information).

However, glycine treatment on alkali-treated jute yarn (ATG) enabled a significant tight packing of the fibre in the jute yarn. As a result, a uniform reduction of yarn diameter (see figure 2(c) and Table S4, Supporting

Information) was observed throughout the length of the yarn, due to the technical fibre fibrillation *via* alkali treatment, and suitable bonding between the functional group of glycine and jute fibres. A scanning electron microscope (SEM) was used to investigate the fibrillar arrangement and surface smoothness of the fibre after alkali and glycine treatments. (figures 2(d)–(f) and S5(b), Supporting Information). Figure 2(d) shows scattered, non-uniform and uncleaned technical fibre surfaces in the UT yarn. The top surface of UT yarn is covered with foreign matters such as oil, waxes, and binding materials such as lignin and hemicelluloses. Alkali treatment removed such impurities and improved the fibre surface roughness [6, 30, 31] (see figure 2(e)). In addition, alkali-treated fibres show grooved appearances which clearly indicates the presence of individual fibre cells (figure 2(e)) and in agreement with previous studies [9, 32]. It could be explained by the fact that hemicelluloses that are located in the inter-microfibrillar region are sensitive to alkali solutions and can easily be removed during the alkali treatment, which causes individual fibre fibrillation in the yarn. The application of glycine on untreated yarn (UTG) improved the connection of technical fibres, figure 2(f). However, glycine treatment on alkali-treated yarns (ATG) increases not only the fibre packing but also the interconnection between the fibrillated fibres created by alkali actions (figure 2(f)). Such improved interconnections create tight fibre packing, and ultimately reduce the yarn diameter similar to the ATG yarn.

3.2. Chemical and thermal characterizations of treated jute yarns

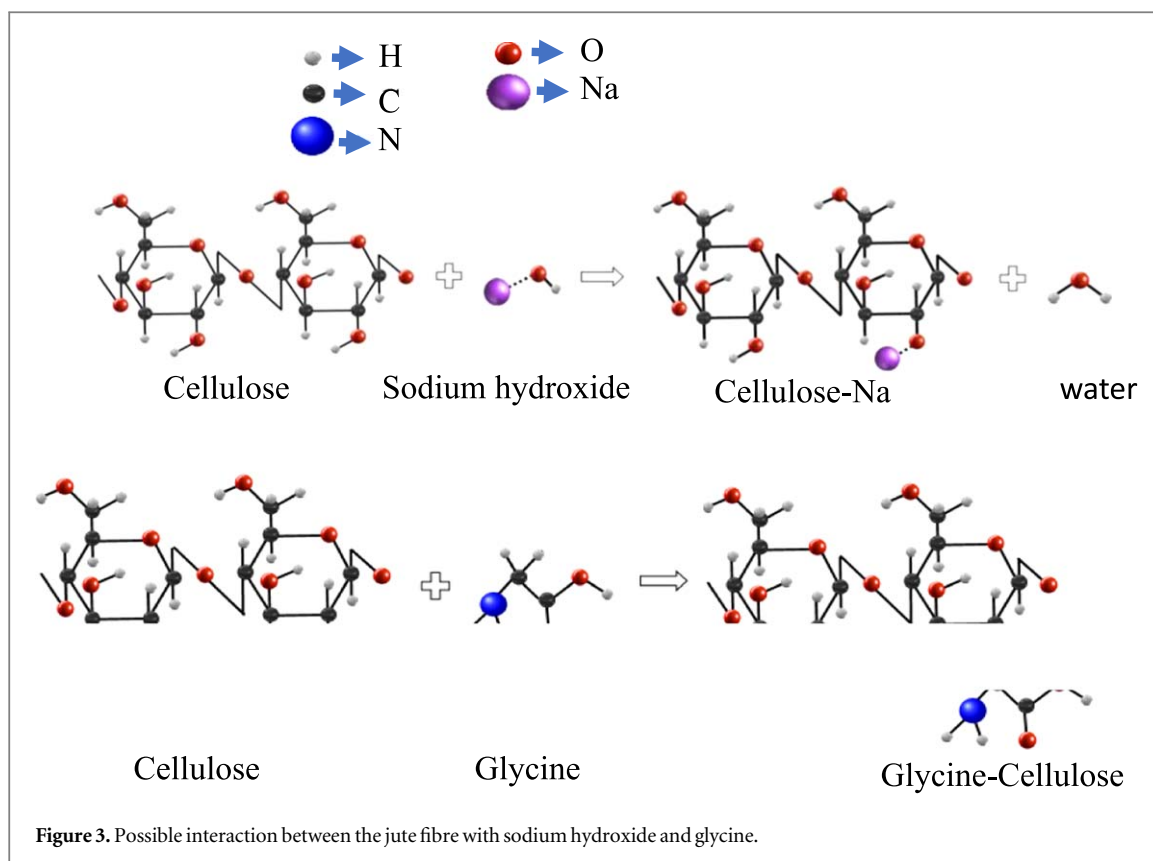
Figure 2(g) shows the FTIR spectra of UT, AT, UTG and ATG jute yarns, which demonstrates four characteristics peaks for UT jute yarns [6, 33]. The peak located at $\sim 3400\text{ cm}^{-1}$ is responsible for the stretching of hydrogen bond, which is originated from the hydroxyl groups present in the cellulose, hemicellulose and lignin of jute fibres. The peaks between 2900 and 2700 cm^{-1} are for the C-H stretching of alkyl groups of cellulose, lignin and hemicelluloses present in jute fibres. In addition, the FTIR spectrum of untreated jute fibres shows peaks at ~ 1738 and $\sim 1249\text{ cm}^{-1}$ band. The peak at $\sim 1738\text{ cm}^{-1}$ is for C=O stretching of carboxylic and ester groups from hemicelluloses presents in the interfibrillar region of UT jute fibres. Furthermore, the band at 1249 cm^{-1} is for the C=O stretching of acetyl groups from lignins of untreated jute fibres. After alkali treatment, these peaks were disappeared which is in agreement with a previous study [6]. The disappearance of such peaks clearly confirms the removal of hemicellulose and lignin from the bundle of technical jute fibres after alkali treatment, which is evident from the optical and SEM images of AT jute yarns (figures 2(b), (e)), and could be explained by the formation of Na-cellulose from the reaction between the cellulose and NaOH, figure 3.

For glycine treatment on UT jute yarn, no additional absorption peaks were observed, which may be due to the presence of impurities in jute fibre that restricts the reaction between the glycine compound and jute fibre. However, ATG jute yarns show slightly extended peaks at $\sim 1540\text{ cm}^{-1}$ and $\sim 570\text{ cm}^{-1}$, which is possibly due to the formation of an amide bond between the amine groups of glycine and carboxylic acid groups of jute fibres. Such observation is further supported by the elimination of the carboxylic ester peak at $\sim 1732\text{ cm}^{-1}$ after glycine treatment. In addition, the peak at 570 cm^{-1} indicates the formation of N=C=O bending [34] again possibly due to the interaction between amine functional groups of glycine and carboxyl groups of jute fibres, (figure 2(g)) Furthermore, the broadening of the peak at $\sim 3200\text{ cm}^{-1}$ may be due to the formation of hydrogen bond between the functional groups of glycine and alkali-treated jute fibres.

The thermal gravimetric analysis (TGA) of UT and treated jute yarns show up to 4%–5% mass loss at $\sim 100^\circ\text{C}$, which indicates the evaporation of water presents in jute fibre (up to 15%) [6]. UT yarns showed two decomposition peaks at $\sim 298^\circ\text{C}$ and $\sim 349^\circ\text{C}$, which are related to the de-polymerization of hemicelluloses, and the decomposition of cellulose after heating, respectively [30]. However, AT jute yarns show a reduction in the onset decomposition temperature to $\sim 272^\circ\text{C}$ and increment in the end-set decomposition to $\sim 356^\circ\text{C}$ (figure 2(h) and Table S3, Supporting Information), maybe due to the removal of polysaccharides, and the increment of crystallinity for the alkali-treated AT jute yarns [6]. In addition, the higher percentage of residues in the AT yarns indicate the improvement of thermal stability of the fibre [35]. Both ATG and UTG yarn show significant improvement in the on-set and end-set decomposition temperature of the jute yarns. For example, ATG yarns exhibit an onset temperature of $\sim 306^\circ\text{C}$ and an end-set decomposition temperature of $\sim 361^\circ\text{C}$ (figure 2(h)). Such improvement in the decomposition temperatures is related to the improvement of crystallinity of jute fibre after the reaction taking place between the glycine and cellulose of jute fibres.

Furthermore, the derivatives of TGA curves for untreated and treated jute yarns show three peaks, where the first peak at 290°C indicates the onset of decomposition of fibre; the second peak at 345°C indicates the decomposition of hemicellulose and celluloses, and the third peak at 440°C indicates the presence of polysaccharides in the fibre (figure 2(i)). After these peaks, the stability in the curve presents the percentage of ash after the decompositions [36]. For ATG yarns all of the three peaks significantly improve which can also be regarded as the reflection of thermal stability improvement of the yarns.

We also performed XPS analysis to quantify the atomic content of carbon, oxygen, nitrogen on the fibre surfaces, and the C/O ratio for different treatment conditions. XPS analysis shows that the untreated yarn (UT)



has a higher C/O ratio of ~ 3.93 (figure 4(a)) due to the abundance of natural binding components such as waxes, lignin and hemicelluloses on the surface of untreated jute fibres. The C/O ratio confirmed the agreement with other natural reinforcing fibres like jute and flax were also found to be 5.45 and 4.03, respectively [6, 37]. After alkali treatment (AT) C/O ratio was reduced slightly to ~ 3.44 due to the decomposition of binding materials (waxes and hemicelluloses) on the jute fibre surface. C/O ratio reduces significantly to ~ 2.36 after glycine treatment (ATG) on alkali-treated jute fibres, which may be due to the increase of oxygen and nitrogen-containing functional groups on glycine-treated jute yarns, figure 4(a).

High-resolution C1s XPS spectra of untreated and treated jute fibres show three main peaks: C-C bond (~ 284.5 eV), C-O epoxy and alkoxy groups (~ 286.4 eV), and C=O carbonyl groups (~ 288 eV), figures 4(b)–(e). The epoxy and alkoxy functional groups were increased significantly after glycine treatment on untreated fibres, which is similar to the results obtained in previous studies [5, 24].

3.3. Tensile properties

Jute yarns, usually brittle, show a sudden decrease in load which corresponds to the failure strain of the yarn as reported by many other researchers who worked with natural fibres [6, 9, 33, 38]. To analyze the tensile behaviour of UT and surface treated-jute yarn, a single yarn tensile test was conducted. Figure 5 shows a large variation in tensile properties which could be due to the variations in the fibre fineness. Therefore, 50 single yarn tests for each type of treated and untreated jute yarns were tested in this work. The values of tensile properties (tensile strength, modulus and strain%) were statistically analyzed using a two-parameter Weibull statistical distribution (Table S6, Supporting Information). Here, we optimize the alkali concentration, the glycine percentage and the effects of pH for surface treatments of jute yarn. These results are listed in (Table S4, Supporting Information). For alkali treatment, 0.5% concentration was found to be the most effective in the case of 10 g l^{-1} glycine concentrations which is considered the best-suited glycine concentration in this study (see Table S4, Supporting Information). The effects of glycine percentage on the alkali-treated jute fibre were studied and optimized as 10 g l^{-1} (Table S4, Supporting Information). The effect of pH on the tensile properties of optimized glycine-treated (10 g l^{-1}) jute yarns were studied in order to understand the intensity of interaction between the glycine moieties and cellulose functional groups at different pH levels. The neutral pH (7) of glycine solutions was found to be good enough to improve the tensile properties of ATG jute yarns (Table S4, Supporting Information). Based on the obtained results, here we used 0.5% alkali concentration and 10 g l^{-1} glycine solution with pH (7) for AT, UTG and ATG yarns. For tensile testing results, UT yarns show relatively lower tensile strength of $\sim 42 \text{ MPa}$, the tensile modulus of $\sim 324 \text{ MPa}$ and tensile strain of $\sim 7.7\%$, figures 5(d)–(f),

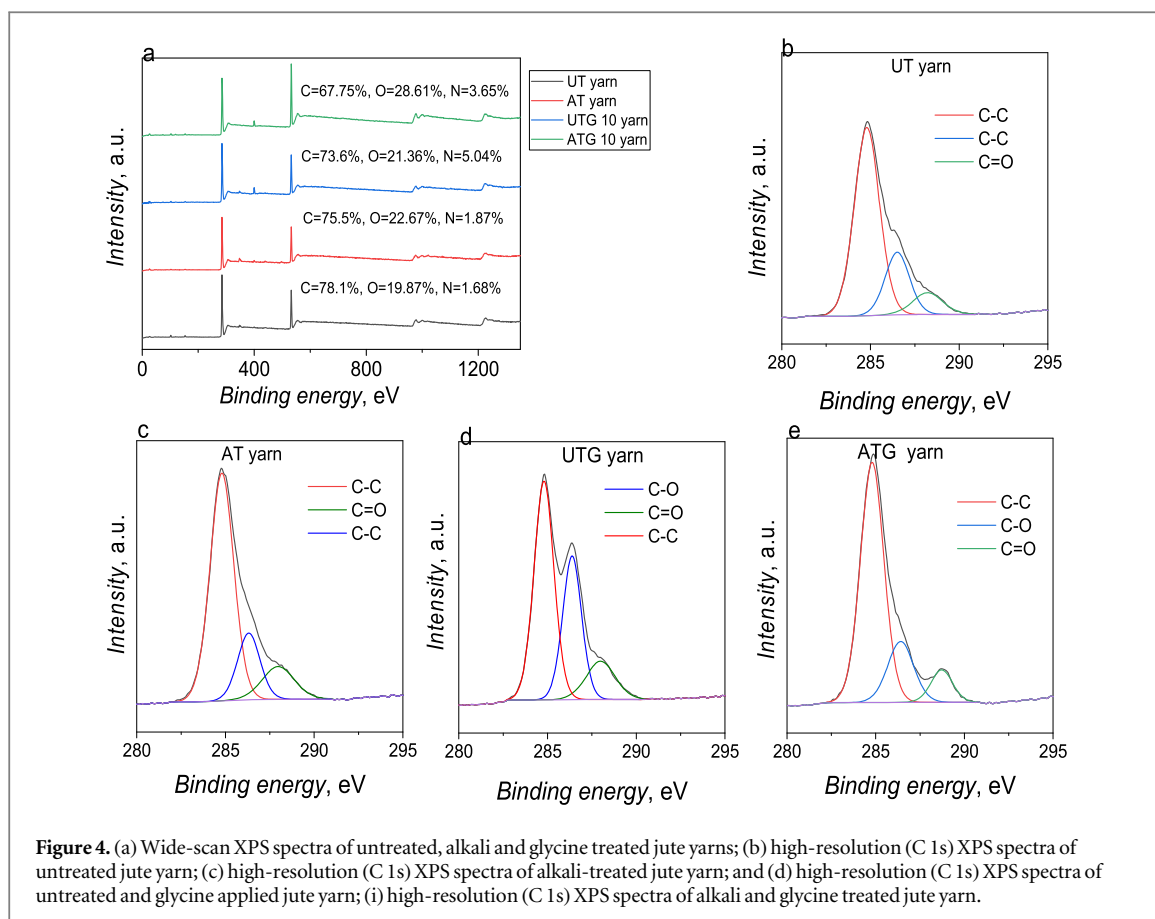
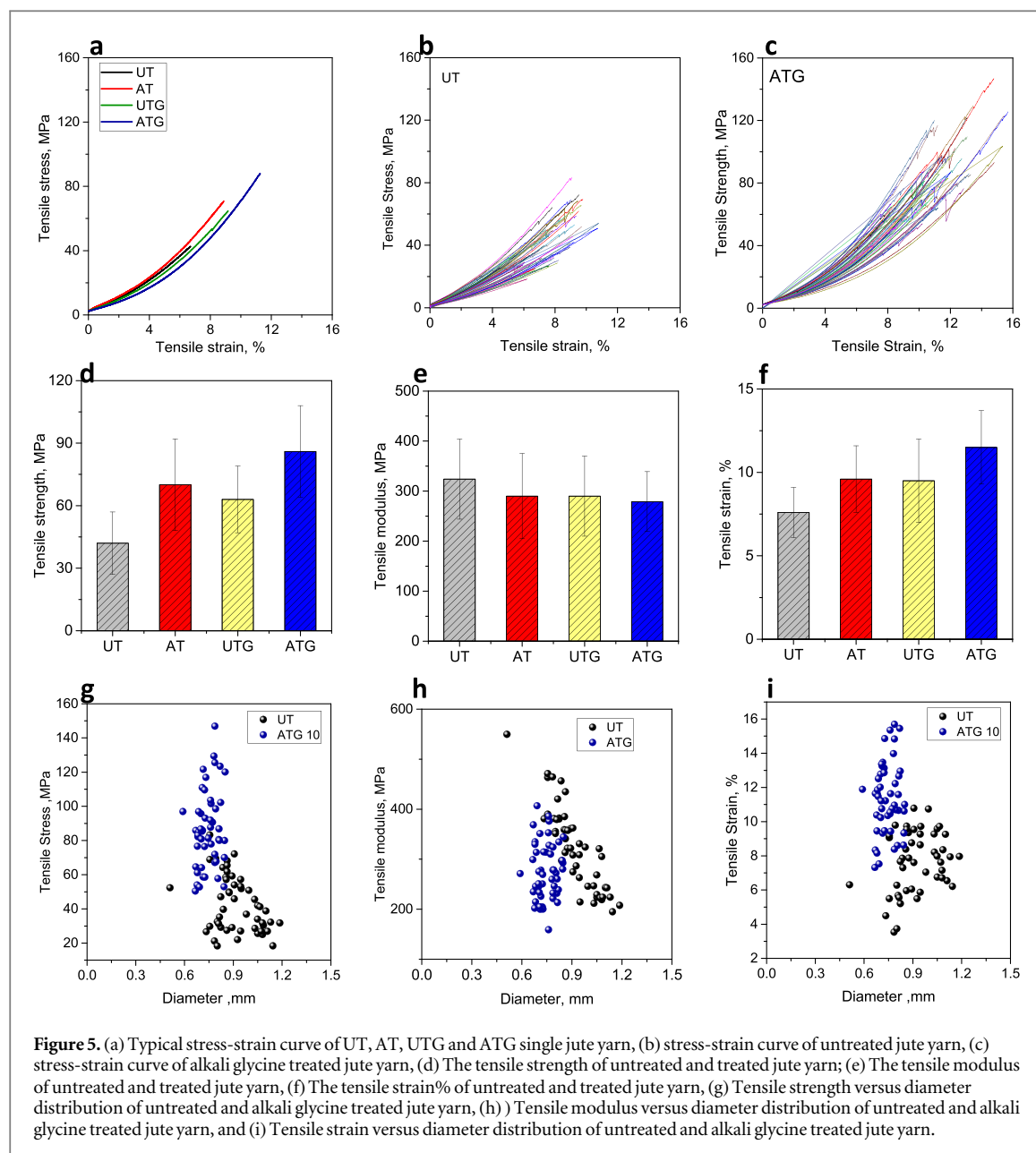


Figure 4. (a) Wide-scan XPS spectra of untreated, alkali and glycine treated jute yarns; (b) high-resolution (C 1s) XPS spectra of untreated jute yarn; (c) high-resolution (C 1s) XPS spectra of alkali-treated jute yarn; and (d) high-resolution (C 1s) XPS spectra of untreated and glycine applied jute yarn; (e) high-resolution (C 1s) XPS spectra of alkali and glycine treated jute yarn.

which is also an agreement with the previous studies related to natural fibres [20, 21]. These lower mechanical properties of UT is mainly due to the waxy cementing layer on the fibre surface composed of low molecular weight fats, lignin, pectin and hemicelluloses [6]. After alkali treatment the values of tensile strength, tensile modulus and tensile strain increase to ~ 70 MPa, ~ 290 MPa and 9.7%, respectively for AT yarns (figure 5(d) and Table S5, Supporting Information). Though no improvement in tensile modulus observed after alkali treatment, however, the tensile strength and strain% increased by $\sim 70\%$ and $\sim 26\%$, respectively, which may be due to the improvement in the packing order of cellulose chains [39]. Alkali solutions are commonly used as a scouring agent in textile processing to remove the impurities from the fibre surfaces and the hemicellulose located in the interfibrillar region of the fibre [6, 33, 39]. As a result, an excessive number of fibrillations is occurred in that region, which enables the improvement in the packing order.

In addition, the orientation of the elementary fibre located in the jute yarn can re-arrange themselves and parallel along the length of the yarn during tensile loading (figure 2(b)). The alkali treatment on natural fibre reduces the spiral shape of cellulose microfibrils that allow the re-arrangement of the cellulose chains and improve the tensile properties of the fibre [39]. Similarly, the alkali treatment of abaca fibre-enabled higher tensile properties than the untreated one, due to the rearrangements of cellulose microfibril along the longitudinal axis [40]. In addition, the alkali treatment can make a better arrangement of the cellulose chain in the fibre which is responsible for the release of internal strain that leads to improving the strength and strain% of jute fibre [41]. Further glycine treatments on alkali-treated jute yarns improve tensile properties significantly (Table S5, Supporting Information), figures 5(d)–(f). The tensile strength and strain% of AT jute yarns increase from ~ 70 MPa and $\sim 0.7\%$ to ~ 86 MPa and 11.5 % for ATG yarns, respectively, which are almost $\sim 105\%$ and $\sim 50\%$ increment in strength and strain values respectively compared to the UT yarn. The enhancement in the tensile strength is supported by the stress-strain curves for UT and ATG yarns in figure 5(b), (c). The possible reason for the improvement in the tensile properties of ATG yarn is related to the strong connection between the AT jute fibres in the yarn with the functional group of glycine *via* suitable chemical or physical bonding. The proposed reaction mechanism is provided in figure 3. In addition, the abundance of oxygen functional group and possible formation of hydrogen bonds have been by XPS and FTIR analysis (figures 4(e) and 2(g)). Thereby, the amorphous region present in the jute fibre cellulose is reduced and more fibrils can pack together to print in the parallel direction to carry more loads. This is in agreement with a previous study [25], where they treated cellulosic cotton fibre with glycine and found a significant increment in the tensile properties due to the bonding



between glycine and cellulosic fibres. In addition, glycine can form zwitterion, which is a compound with no electrical charge since it has both positively and negatively charged functional groups. Interaction between the glycine moieties with the hydroxyl groups of jute fibre takes place in the amorphous region of fibre which renders in absorbing more energy during the mechanical loading of the fibres which ultimately improves the overall tensile properties of ATG yarn [25, 41].

The improved mechanical properties could also be described based on the SEM image observations found in figure 2(f) of this study where an excellent fibre packing and the paralleled, fibrillated fibres interconnection were achieved for ATG yarns. As a result, there was no stress decay to print the fibre in the parallel direction leading to improve stress carrying capacity of ATG yarn during tensile loading applications. Between ATG and UTG yarns, ATG yarns showed better tensile strength and strain properties, because ATG yarns contain more hydroxyl groups for the alkali treatment which enables better interaction with the functional groups of glycine compared to UTG yarns. However, ATG yarns exhibited a lower tensile modulus value, this is possibly due to the new bonds formed in the ATG yarns which increases the fibrillar cohesion to enhance the strain with respect to the increase of stress (see figure 5(e)). Stress-strain curves of ATG yarn showed more improvement compared to UT yarns (see figure 5(c)).

Here, improvement in tensile properties of jute yarn is related to the better packing of fibrils in the fibre due to strong chemical interactions after glycine treatments. Besides this, we observe diameter of the yarns have changed after glycine treatment which also resulted in improving tensile properties of jute yarn is also shown in

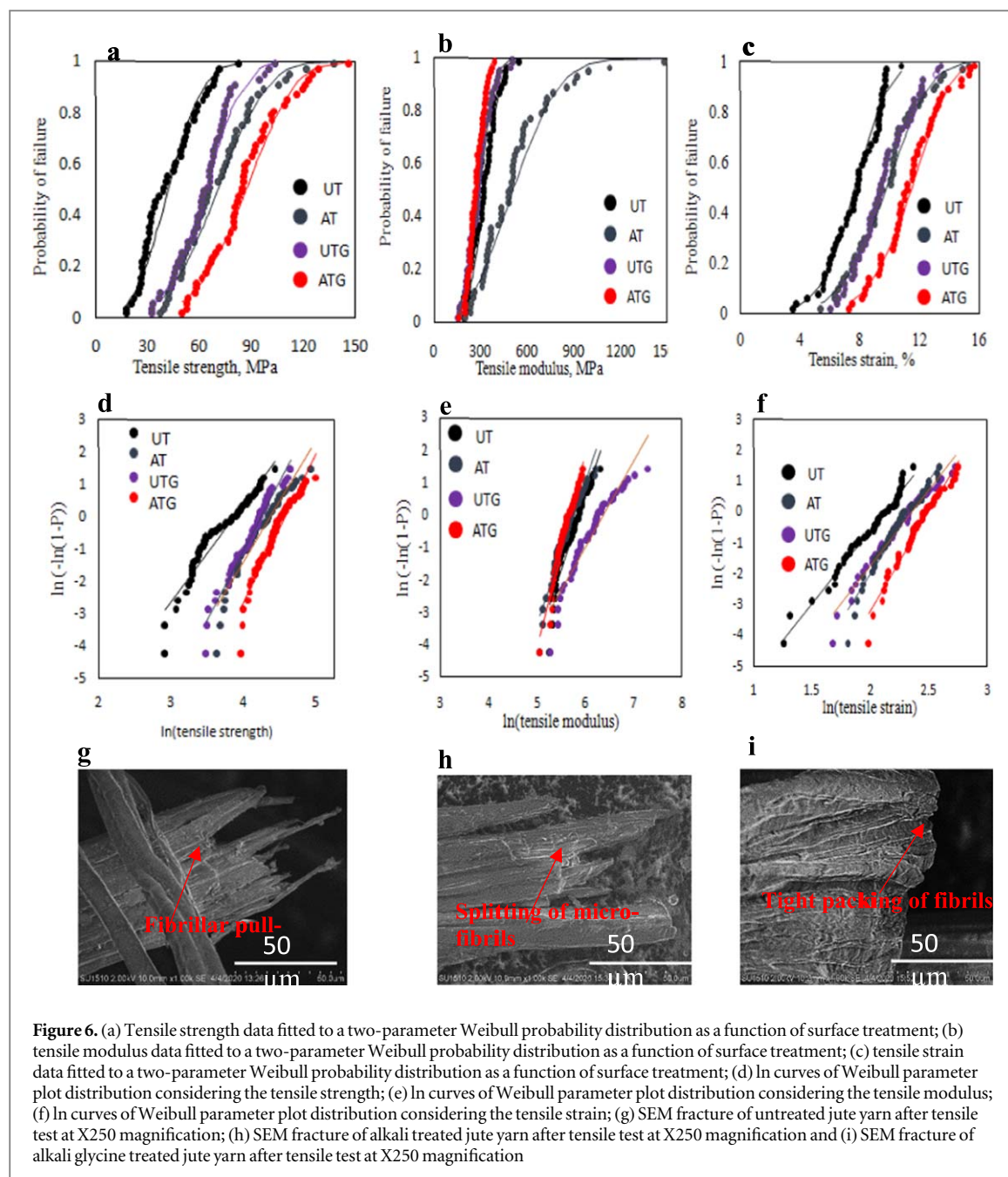


Figure 6. (a) Tensile strength data fitted to a two-parameter Weibull probability distribution as a function of surface treatment; (b) tensile modulus data fitted to a two-parameter Weibull probability distribution as a function of surface treatment; (c) tensile strain data fitted to a two-parameter Weibull probability distribution as a function of surface treatment; (d) \ln curves of Weibull parameter plot distribution considering the tensile strength; (e) \ln curves of Weibull parameter plot distribution considering the tensile modulus; (f) \ln curves of Weibull parameter plot distribution considering the tensile strain; (g) SEM fracture of untreated jute yarn after tensile test at X250 magnification; (h) SEM fracture of alkali treated jute yarn after tensile test at X250 magnification and (i) SEM fracture of alkali glycine treated jute yarn after tensile test at X250 magnification

figures 5(g)–(i). This result can be supported from the SEM and optical image observations (figure 2(c) and (f)) that yarn with a smaller diameter have reduced porosity and fewer impurities in the fibre. We conduct statistical analysis to validate the data obtained after tensile experiments (see Table S6, Supporting Information). Weibull statistical distribution was performed to evaluate the scale parameter (α) and shape parameter (β) of the scattered tensile results

Here α predicts the experimental results and β indicates the modulus of Weibull distribution mainly known as Weibull modulus. Higher the value in Weibull modulus better the improvement in the scattering effect in the results. Figures 6(a)–(f) shows the Weibull distribution for tensile strength, modulus and strain of jute yarns as the probability of failure (figures 6(a)–(c)) and \ln curve (figures 6(d)–(f)). It is seen that this statistical model provided an excellent fitting of the data for the tensile properties of the yarns. Moreover, Weibull distribution calculated a reasonable numerical prediction of the experimental data, provided in (Table S6, Supporting Information). It was found that tensile strength and strain values (scale parameter) of the yarn were improved after introducing the alkali and glycine treatments on jute yarn (ATG). In this case, both the probability of failure and \ln curves were seen to shift from left to right significantly when ATG yarns were compared with UT jute yarns. The Weibull modulus was obtained from the \ln curve of the untreated and treated yarns as shown in (Table S6, Supporting Information). UT yarn showed relatively lower value in Weibull modulus ($\sim 3, 4.5$ and 4.8 ,

for tensile strength, tensile modulus and tensile strain respectively) due to the high scattering of UT yarns linked to the presence of impurities in the fibre located in the yarn. However, the Weibull modulus was increased to 6.1 for tensile strength, 5.9 for tensile modulus and 6.1 for failure strain of the ATG jute yarn which could be because of the better bonding between the jute fibre and the glycine, as we discussed in the earlier sections of this work. The higher value in Weibull modulus for the jute yarn was found in similar to synthetic fibres reported in the literature. Chawla *et al* [42] experimented with the value of 4.6 for the Weibull modulus of ceramic fibre, whereas this study calculated the Weibull modulus of 6.1 of ATG yarns which confirmed the weakest link in the fibre caused by flaws present in the fibre was reduced significantly after introducing alkali and glycine treatment on jute yarns.

3.4. Fractographic study of jute fibre yarn

We investigated the fracture specimen of different treated jute yarns using SEM. In this investigation, we observed that bundles of micro-fibrils are present in the yarn which can be seen in figures 6(g)–(i). In the case of the broken specimen from UT yarn, a very uneven fracture of jute fibre bundles with fibre pull-out from the skin of UT yarn is visible in figure 6(g). This might be due to the presence of impurities into the interfibrillar network of UT yarn which is also supported by other studies [6, 43]. Fibre splitting was observed as the dominant fracture feature with a small amount of fibre pull-out for AT yarns (see figure 6(h)). The dominance of fibre splitting might be related to the improvement in the crystallinity of AT jute fibres and removal of the hemicelluloses which act as the stress concentration points of jute fibres. The brittle fracture occurred in the transverse direction of the UTG yarns to some extent for the glycine treatment whereas in (figure S6, Supporting Information). ATG yarn showed a vivid brittle fracture surface without any fibre pull-out (see figure 6(i)). For ATG yarns, fibrils in the yarn were broken in the same order along the transverse direction indicating that the improved packing of microfibrils created with both alkali and glycine treatments evenly distributed along the length of the fibre.

3.5. Comparative study with the literature

A comparison was tried to make in (Table S7, Supporting Information) for the tensile properties particularly on tensile strength and strain observed between this study and other natural fibre based (flax, hemp and jute) yarns studies reported in the literature. The studies of jute yarn mechanical properties are very limited in the literature. A direct comparison is difficult as the experimental conditions in those studies are different from this study. In addition, different fibres have different constituents' ratios, which have a direct impact on the mechanical properties. As found in the comparison, the alkali glycine treated jute yarns (ATG) showed an excellent and huge improvement both in tensile strength and strain properties in this study compared to any other reported tensile properties of jute yarns found in the literature.

4. Conclusions

In this study, aqueous glycine treatment was applied on untreated and alkali-treated jute yarns, and their influences on the chemical, thermal, morphological and mechanical properties of jute yarn were evaluated. The results indicate an extremely positive effect of glycine treatment on jute yarn towards structural properties improvement. Glycine treatment on alkali-treated jute yarns (ATG) brought a remarkable improvement by almost ~105% and ~50 % increment in tensile strength and strain properties respectively compared to untreated jute yarns. The significant improvements in the mechanical properties of newly developed glycine and alkali-treated jute yarns (ATG) achieved in this work will be helpful to develop the use of jute yarn-based woven or multi-axial textile architectures as reinforcing elements in manufacturing natural plant-based composites for structural applications.

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Data availability statement

The data that support the findings of this study are openly available at the following URL/DOI: <https://doi.org/10.21203/rs.3.rs-151073/v1>.

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References

- [1] Karim N, Zhang M, Afroj S, Koncherry V, Potluri P and Novoselov K S 2018 Graphene-based surface heater for de-icing applications *RSC Adv.* **8** 16815–23
- [2] Pickering K L, Efendy M G A and Le T M 2015 A review of recent developments in natural fibre composites and their mechanical performance *Compos. Part A Appl. Sci. Manuf.* **83** 98–112
- [3] Karim M N, Rigout M, Yeates S G and Carr C 2014 Surface chemical analysis of the effect of curing conditions on the properties of thermally-cured pigment printed poly (lactic acid) fabrics *Dye. Pigment.* **103** 168–74
- [4] Karim M N, Afroj S, Rigout M, Yeates S G and Carr C 2015 Towards UV-curable inkjet printing of biodegradable poly (lactic acid) fabrics *J. Mater. Sci.* **50** 4576–85
- [5] Nahar K, Arju S N, Ferdush J, Islam M and Akter T 2020 Colorimetric analysis and fastness properties of jute fabric dyed with eucalyptus leaves *Tekstiles* **63** 195–202
- [6] Sarker F, Karim N, Afroj S, Koncherry V, Novoselov K S and Potluri P 2018 High-Performance Graphene-Based Natural Fiber Composites *ACS Appl. Mater. Interfaces* **10** 34502–12
- [7] Defoirdt N et al 2010 Assessment of the tensile properties of coir, bamboo and jute fibre *Compos. Part A Appl. Sci. Manuf.* **41** 588–95
- [8] Hearle J W S 1963 The fine structure of fibers and crystalline polymers. I. Fringed fibril structure *J. Appl. Polym. Sci.* **7** 1175–92
- [9] Mukherjee A, Ganguly P K and Sur D 2008 Structural Mechanics of Jute: the effects of hemicellulose or lignin removal *J. Text. Inst.* **84** 348–53
- [10] Saha P, Manna S, Chowdhury S R, Sen R, Roy D and Adhikari B 2010 Enhancement of tensile strength of lignocellulosic jute fibers by alkali-steam treatment *Bioresour. Technol.* **101** 3182–7
- [11] Gassan J and Bledzki A K 1999 Possibilities for improving the mechanical properties of jute/epoxy composites by alkali treatment of fibres *Compos. Sci. Technol.* **59** 1303–9
- [12] Arfaoui M A, Dolez P I, Dubé M and David E 2017 Development and characterization of a hydrophobic treatment for jute fibres based on zinc oxide nanoparticles and a fatty acid *Appl. Surf. Sci.* **397** 19–29
- [13] Militký J and Jabbar A 2015 Comparative evaluation of fiber treatments on the creep behavior of jute/green epoxy composites *Compos. Part B Eng.* **80** 361–8
- [14] Sarker F, Potluri P, Afroj S, Koncherry V, Novoselov K S and Karim N 2019 Ultrahigh Performance of Nanoengineered Graphene-Based Natural Jute Fiber Composites *ACS Appl. Mater. Interfaces* **11** 21166–76
- [15] Dang C Y et al 2019 Enhanced interlaminar shear strength of ramie fiber /polypropylene composites by optimal combination of graphene oxide size and content *Compos. Part B Eng.* **168** 488–95
- [16] Khondker O A, Ishiaku U S, Nakai A and Hamada H 2005 Tensile, flexural and impact properties of jute fibre-based thermosetting composites *Plast. Rubber Compos.* **34** 450–62
- [17] De Albuquerque A C, Joseph K, Hecker De Carvalho L and D'Almeida J R M 2000 Effect of wettability and ageing conditions on the physical and mechanical properties of uniaxially oriented jute-roving-reinforced polyester composites *Compos. Sci. Technol.* **60** 833–44
- [18] Sarkar F, Akonda M and Shah D U 2020 Mechanical properties of flax tape-reinforced thermoset composites *Materials (Basel)*. **13** 1–16
- [19] Karim N, Sarker F, Afroj S, Zhang M, Potluri P and Novoselov K S 2021 Sustainable and Multifunctional Composites of Graphene-Based Natural Jute Fibers *Adv. Sustain. Syst.* **5** 1–12
- [20] Sharif Ullah A M M, Shahinur S and Haniy H 2017 On the mechanical properties and uncertainties of jute yarns *Materials (Basel)*. **10** 450
- [21] Bensmail Y, El moumen A, Lmai F and Imad A 2019 The effects of the temperature on the mechanical properties of natural yarns *MATEC Web Conf.* **286** 3003
- [22] Foruzanmehr M, Vuillaume P Y, Robert M and Elkoun S 2015 The effect of grafting a nano-TiO₂ thin film on physical and mechanical properties of cellulosic natural fibers *Mater. Des.* **85** 671–8
- [23] Perremans D, Hendrickx K, Verpoest I and Van A W 2018 Vuure, 'Effect of chemical treatments on the mechanical properties of technical flax fibres with emphasis on stiffness improvement *Compos. Sci. Technol.* **160** 216–23
- [24] Li Y, Chen C, Xu J, Zhang Z, Yuan B and Huang X 2015 Improved mechanical properties of carbon nanotubes-coated flax fiber reinforced composites *J. Mater. Sci.* **50** 1117–28
- [25] Remadevi R, Gordon S, Wang X and Rajkhowa R 2018 Tensile, physical, and microstructure properties of glycine treated cotton fibers *Text. Res. J.* **88** 1356–1366
- [26] Bose S, Kuila T, Mishra A K, Kim N H and Lee J H 2012 Dual role of glycine as a chemical functionalizer and a reducing agent in the preparation of graphene: An environmentally friendly method *J. Mater. Chem.* **22** 9696–703
- [27] Kim J T and Netravali A N 2011 Development of aligned-hemp yarn-reinforced green composites with soy protein resin: Effect of pH on mechanical and interfacial properties *Compos. Sci. Technol.* **71** 541–7
- [28] Wang F, Shao J, Keer L M, Li L and Zhang J 2015 The effect of elementary fibre variability on bamboo fibre strength *Mater. Des.* **75** 136–42
- [29] Fernandes E M, Mano J F and Reis R L 2013 Hybrid cork-polymer composites containing sisal fibre: Morphology, effect of the fibre treatment on the mechanical properties and tensile failure prediction *Compos. Struct.* **105** 153–62
- [30] Orue A, Jauregi A, Peña-Rodríguez C, Labidi J, Eceiza A and Arbelaz A 2015 The effect of surface modifications on sisal fiber properties and sisal/poly (lactic acid) interface adhesion *Compos. Part B Eng.* **73** 132–8
- [31] Stocchi A, Lauke B, Vázquez A and Bernal C 2007 A novel fiber treatment applied to woven jute fabric/vinylester laminates *Compos. Part A Appl. Sci. Manuf.* **38** 1337–43
- [32] Mwaikambo L Y 2009 Tensile properties of alkalis jute fibres *BioResources* **4** 566–88

- [33] Roy A, Chakraborty S, Kundu S P, Basak R K, Majumder S B and Adhikari B 2012 Improvement in mechanical properties of jute fibres through mild alkali treatment as demonstrated by utilisation of the Weibull distribution model *Bioresour. Technol.* **107** 222–8
- [34] Saroj A L, Singh R K and Chandra S 2013 Studies on polymer electrolyte poly(vinyl) pyrrolidone (PVP) complexed with ionic liquid: Effect of complexation on thermal stability, conductivity and relaxation behaviour *Mater. Sci. Eng. B Solid-State Mater. Adv. Technol.* **178** 231–8
- [35] Kandola B, Sarker F, Luangtriratana P and Myler P 2016 Thermal protection of carbon fiber-reinforced composites by ceramic particles *Coatings* **6** 22
- [36] Müssig J and Haag K 2015 Biofiber reinforcements *Composite Materials* (Amsterdam: Elsevier)
- [37] Le Duigou A, Kervoelen A, Le Grand A, Nardin M and Baley C 2014 Interfacial properties of flax fibre–epoxy resin systems: existence of a complex interphase *Compos. Sci. Technol.* **100** 152–7
- [38] Fiore V, Scalici T, Nicoletti F, Vitale G, Prestipino M and Valenza A 2016 A new eco-friendly chemical treatment of natural fibres: Effect of sodium bicarbonate on properties of sisal fibre and its epoxy composites *Compos. Part B Eng.* **85** 150–60
- [39] Sawpan M A, Pickering K L and Fernyhough A 2011 Effect of various chemical treatments on the fibre structure and tensile properties of industrial hemp fibres *Compos. Part A Appl. Sci. Manuf.* **42** 888–95
- [40] Cai M, Takagi H, Nakagaito A N, Li Y and Waterhouse G I N 2016 Effect of alkali treatment on interfacial bonding in abaca fiber-reinforced composites *Compos. Part A Appl. Sci. Manuf.* **90** 589–97
- [41] Bledzki A K and Gassan J 1999 Composites reinforced with cellulose based fibres *Progress in Polymer Science (Oxford)* **24** 221–74
- [42] Chawla N, Kerr M and Chawla K K 2005 Monotonic and cyclic fatigue behavior of high-performance ceramic fibers *J. Am. Ceram. Soc.* **88** 101–8
- [43] Aziz S H and Ansell M P 2004 The effect of alkalization and fibre alignment on the mechanical and thermal properties of kenaf and hemp bast fibre composites: Part 1 - polyester resin matrix *Compos. Sci. Technol.* **64** 1219–30